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Validation of 3 years of ozone measurements over Network for the Detection of Stratospheric Change station Lauder, New Zealand

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Abstract. A large number of ozone profiles measured by using various methods (lidar, ozonesondes, microwave radiometer, and Stratospheric Aerosol and Gas Experiment II) over Lauder, New Zealand, between late 1994 and early 1998 are intercompared. These profiles are also used to validate a collocated Dobson spectrophotometer. Between March 1996 (August 1996 for sonde measurements) and January 1998, all instruments were operational and no instrument changes took place. The ozone number densities averaged over this period agreed within 5% in the 20- to 35-km range. Between 12 and 20 km, lidar and sonde results deviated by less than 8%, and lidar and SAGE II results deviated by less than 15%. The ozone column densities measured by the Dobson spectrophotometer agreed within 3% with the integrated ozone profiles. The various methods are discussed, and modifications are proposed. They comprise a decrease of the reported sonde altitudes of about 125 m (±50 m) to correct for the response time of the sonde's chemical solution, the use of more accurate molecular parameter values in the lidar algorithm, and a 2.5% decrease in SAGE II ozone densities. Improved agreement between the average ozone profiles to within 1.5% for lidar and sondes (20-35 km) and within 2.5% for lidar and SAGE II (20-35 km) is achieved. The Dobson results are found to be influenced by the annual cycle of the temperature profile through the altitude-averaged ozone absorption cross section, which has been assumed to be constant in the presented data but actually varies with an amplitude of 2% over Lauder.

1. Introduction

Altitude profiles of the ozone number density ("ozone profiles") have been and are being measured at several stations of the Network for the Detection of Stratospheric Change (NDSC), in order to detect stratospheric trends and to provide a calibration for satellite instruments. Accurate detection of trends in the ozone profiles is needed for the assessment of the influence of anthropogenic sources, such as chlorofluorocarbons (CFCs), on the ozone layer [*Stratospheric Processes and Their Role in Climate (SPARC)*, 1998]. Only the combination of measurements at high-quality ground-based stations and satellite instruments with global coverage may provide insight into environmental developments on a global scale.

Trends in stratospheric ozone concentrations over several sites have been investigated in the framework of a worldwide project [*SPARC*, 1998], mainly using sondes and satellite instruments. Because of natural variations, regular ozone profile measurements spanning about 20 years or longer are needed to extract ozone trends. To enable trend detection at the earliest possible stage, it is imperative that the ozone profiles are sufficiently accurate in comparison with natural variations expected. Also the instrument stability should be high, because drift may enhance or obscure atmospheric trends.

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The quality of measurements at NDSC sites is investigated regularly, during short-term campaigns [e.g., *McDermid et al.*, 1998a, b;*McPeters et al.*, 1999;*Steinbrecht et al.*, 1999] as well as through longer-term validations [e.g., *Tsou et al.*, 1995]. In these investigations, measurements employing different techniques are compared. Intercomparison of the different techniques permits study of sensitivities for different systematic effects, e.g., interference from the presence of aerosols or clouds. Apart from providing a validation, intercomparisons also often lead to improvements in the individual results, thereby reducing inaccuracies. Furthermore, if the differences between ozone profiles measured by the various instruments are sufficiently small, then combination of these profiles is useful. Since results discussed in this work are complementary both in the altitude regions that are covered and in the number of measurements per period of time, their future combination will result in a more extensive ozone time series, which may allow for earlier trend detection than that provided by single instruments.

Although measurements of the ozone column density do not provide the detailed trend information that is provided by series of profiles, they do contribute to knowledge of the global development of the ozone layer. Such measurements have taken place with Dobson spectrophotometers since the 1920s. Presently, a global network of over a hundred Dobson spectrophotometers is operational, and regular calibration through comparison to a standard Dobson instrument [e.g., *Komhyr et al.*, 1989] takes place in most cases. Since Dobson spectrophotometers are so widespread, critical validation of their results is useful.

The current work provides a validation of several years of ozone measurements performed over NDSC station Lauder, New Zealand (45° S, 170° E). All Lauder-based instruments that measure ozone profiles as well as the satellite-borne Stratospheric Aerosol and Gas Experiment II (SAGE II) are intercompared. Ozone profiles have been measured at Lauder since August 1986 by using electrochemical concentration cell (ECC) ozonesondes. More recently, a microwave radiometer (1992) and a differential absorption lidar (DIAL) system (late 1994) were added. SAGE II measurements commenced in October 1984. Also ozone column densities measured with a Dobson spectrophotometer, which has been operated at Lauder since January 1987, are validated by intercomparison with the integrated results of the profiling instruments.

Ozone profiles measured by the various instruments at Lauder were investigated previously, during the 2-week Ozone Profiler Assessment at Lauder (OPAL) campaign [*McDermid et al.*, 1998a, b] in April 1995. The single ozone profiles measured by the Lauder sondes and stratospheric lidar and by a traveling standard lidar operated by the Goddard Space Flight Center agreed within 5% for the 15- to 42-km region (32 km for sondes). Above 42 km, the lidar results were unreliable. Microwave ozone densities were systematically higher (by about 5%) than those of the other discussed instruments in the region from 22 to 42 km [*McDermid et al.*, 1998b].

This paper has several objectives. First, provide an improved validation of the existing ozone measurements by extending the 2-week OPAL campaign and using more measurements over a longer time period (3 years) including all seasons and a wider variety of meteorological and other conditions. The study also reflects hardware changes made after OPAL. Therefore the results presented here give a more representative validation of the Lauder ozone profiles than the OPAL campaign did (NDSC data are available from http://www.ndsc.ncep.noaa.gov/).

Second, modifications to the lidar, SAGE II and sonde analyses are proposed which improve the agreement between the ozone profiles. We emphasize that these modifications emerged from a discussion of the discrepancies between the various data sets. This provides an additional rationale for performing multiyear intercomparisons.

And finally, the 3-year intercomparison allows us to draw conclusions about instrument stabilities and thus about the limitations of the detection of trends. Also the effects of trends other than in ozone density itself can be studied. As an illustration, we show that changes in temperature influence the value of the altitude-averaged ozone absorption cross section, which for the Dobson spectrophotometer results is assumed to be constant over time. This assumption influences the assessment of a trend in the ozone concentration.

2. System Descriptions

In this section, system descriptions are presented, along with discussions of the measurement accuracies. For all systems except the lidar, these descriptions are succinct, and references are given to articles that contain more detailed information.

During the operation of the various instruments, comparisons of the individual profiles have been made on a regular basis. Also the integrated profiles were regularly compared with the Dobson ozone column densities. However, it is emphasized that the instruments operated completely independently, without adaptation of results to achieve better agreement. In particular, the sonde ozone profiles were not scaled to agree with the Dobson results, which is common practice at many other sites.

We will define the accuracy of a measurement as the sum of its repeatability (often referred to as precision) and its systematic errors. All ozone profiles discussed in this paper are provided with accuracy estimates, with the exception of the RIVM lidar results which contain estimates of the repeatability only.

2.1. Differential Absorption Lidar System

The lidar ozone profiles discussed in this paper were measured by the stratospheric lidar operated at Lauder by the Dutch Institute for Public Health and the Environment (RIVM). A detailed description of the RIVM stratospheric lidar system was given by *Swart et al.* [1995].

Ozone profiles are derived by using the differential absorption lidar method (see, e.g., *Measures* [1984]). Laser pulses at two wavelengths (308 and 353 nm), only one of which is affected by ozone absorption, are sent into the atmosphere. Two slightly different DIAL methods can be distinguished. The elastic DIAL method uses signals detected after elastic scattering in the atmosphere, whereas the inelastic DIAL method employs signals returned by (inelastic) vibrational Raman scattering by nitrogen molecules. In both cases, the ozone profile is derived from the ratio of the two detected signals, after a correction has been made for the differences in atmospheric transmission (due to the wavelength dependence of Rayleigh scattering). During periods with a high aerosol number density the elastic method, in which signals are detected from scattering by molecules as well as by aerosols, breaks down. The inelastic method, however, still yields valid results [*McGee et al.*, 1993]. During the period in which the lidar measurements were performed, the aerosol concentration was sufficiently low for the elastic method to be used. For technical reasons, the inelastic method was sometimes used below 20 km. Only the elastic DIAL equation will be discussed here, and the assumption that the influence of aerosol scattering is sufficiently small will be made. The elastic DIAL equation is usually expressed as [*McDermid et al.*, 1990]

$$n_{O_3}(r) = -\frac{1}{2\sigma_{O_3}(r)}\left[\frac{d}{dr}\ln\frac{S_{308}(r)}{S_{353}(r)}\right] - \frac{n(r)}{\sigma_{O_3}(r)}\left[\sigma_{R,308}(r) - \sigma_{R,353}(r)\right]. \quad (1)$$

Here r is the geometric altitude, n_{O_3} is the ozone number density, σ_{O_3} is the differential ozone absorption cross section between 308 and 353 nm, n is the air number density, S is the background-corrected return signal, and σ_R is the Rayleigh extinction cross section. S and σ_R are subscripted with the wavelength.

The DIAL method is self-calibrating, i.e., constants such as system efficiency and extinction in the troposphere will vanish.

There is a variety of methods for establishing the first term on the right-hand side of equation (1) for a specific altitude r_i, each with its own resolution and repeatability [*Beyerle and McDermid*, 1999]. In the RIVM lidar case, a linear fit is performed through the logarithm of the signal ratio at $2N + 1$ altitude bins, described by

$$\ln\frac{S_{308}(r_j)}{S_{353}(r_j)}, \quad (2)$$

with $j = [i - N, i + N]$. Here, r_j are altitudes from the collection of altitude bins within a distance of N times the altitude bin size from r_i, where N is an integer. The slope of this linear fit, divided by $-2\sigma_{O_3}(r_i)$, yields the required term at altitude r_i. The signal-to-noise ratio of the detected signals decreases with increasing altitude, and to get sufficiently accurate fits, N is chosen to be increasing with altitude, so that typical intervals over which the fits are made range from 2.1 to 4.8 km. We will adopt half of the length of such an interval as the resolution of the lidar measurement (see *Beyerle and McDermid* [1999] for a discussion of different definitions of lidar ozone profile resolutions). The measurement repeatability is defined as the statistical error of the performed linear fit.

In the stratosphere, the first term on the right-hand side of equation (1) is large in comparison with the applied Rayleigh correction (the second term). The Rayleigh correction, which decreases with increasing altitude, is determined from molecular parameters and a sonde air density profile. The same sonde also provides a temperature profile, which is used to determine the temperature dependent ozone absorption cross section. A small interdependence of ozone profiles measured by sonde and lidar is introduced in this way, as the conversion from sonde ozone partial pressure to ozone number density is performed by using the same (sonde) density and temperature data, but this is assumed to be negligible.

After November 1994, when measurements at Lauder commenced, several changes have been made to the lidar hardware. First, the measurement domain was increased from 100 km originally to 150 km (early April 1995) and 300 km (April 1995), enabling better fits to the background light levels (which contain a contribution due to signal-induced noise of the detectors). This implied a corresponding change in the resolution with which the altitudes were measured from 100 to 150 and 300 m. Second, deterioration of the laser beam divergence during early 1995 made it necessary to increase the detector field of view from 0.6 to 0.8 mrad (November 1995). However, problems remained as some of the light collected by the telescope extended beyond the size of the detectors, leading to invalid ozone values below a certain altitude. This problem was resolved by adding a field lens in the detector section of the lidar (March 1996). Therefore the part of the ozone profile obtained at low altitudes before March 1996 must be treated with caution (see subsection 4.1). Also changes to the processing software were made; the data currently in the NDSC database were all processed with software version 7.3.

The geometric altitude in the lidar ozone profiles is determined by measurement of the delay between the

emission of a light pulse from the laser and the detection of the atmospheric return signal. Except for a small altitude offset, caused by the time difference between the start of counting by the detectors and the emission of the laser pulse, the altitudes are determined well, to within about 10 m. The altitude offset is measured about once a year and depends on details of the laser configuration. The largest influence on the observed altitude offset is the laser temperature, which under typical operating conditions introduces a fluctuation of about 10 m. An investigation of the analysis software shows that the value used for the time difference was measured under atypical conditions. The altitude scale derived differs by about 50 m from that derived under more representative conditions, and therefore an altitude increase of 50 m to all lidar ozone profiles must be applied (see subsection 5.1). In future, the use of an optical trigger in the lidar system is intended, which will remove the described altitude determination problem.

A brief evaluation of the systematic errors of the RIVM results is given here. At altitudes where the strength of the background signal is negligible in comparison with the strength of the atmospheric return signal (typically, below 40 to 45 km), two main sources of systematic errors can be distinguished. First, uncertainties in the Rayleigh correction, which are in turn due to uncertainties in the density profiles used, may contribute to the inaccuracy of the lidar ozone profile. The Rayleigh correction is proportional to the air density and is typically less than 1% above 15 km [*Mégie and Menzies*, 1980] when density profiles are recorded within a few days from the lidar measurement. However, in the lidar data (version 7.3), sonde density profiles were used that were recorded within a week, typically. This occasionally led to errors in the ozone density of up to 3% at 15 km. Above 30 km, the contribution to the overall lidar accuracy is negligible.

Second, the extracted lidar ozone number densities are inversely proportional to the ozone cross section, which is temperature dependent. Its accuracy is predominantly determined by the accuracy of the temperature profile used. By comparing the sonde temperature profiles with the National Centers for Environmental Prediction (NCEP) data that were recorded less than a few hours apart from the lidar measurements, typical differences in the extracted ozone cross sections of about 2% were found, and they were neither systematically positive nor negative. This provides an order of magnitude estimate of the reliability of the ozone cross section profiles used.

In conclusion, by using the lidar repeatabilities only, as is done in this work, two systematic error sources are neglected. The first relates to variations in the atmospheric density and is less than 1% above 15 km. The second error source relates to fluctuations in the atmospheric temperature profiles and is about 2% at all altitudes, on average, but will be reduced in the future by the application of an improved lidar data analysis.

An algorithm intercomparison between several NDSC lidar groups [*Godin et al.*, 1999] showed that the ozone profiles agree well but that the repeatability estimates reported by RIVM (and the York University group) are too conservative in two altitude regions (typically, 15-18 and 23-28 km). Further investigation by us has shown that the actual repeatability in these altitude regions is 2 to 3 times better than reported. The discrepancy stems from using the standard deviation of a linear rather than a second-order fit to the DIAL curve in order to determine the ozone profile repeatability. Since below 28 km the lidar repeatability is relatively good in comparison with the accuracy of other instruments, a change of the reported lidar repeatability will not significantly influence the combined accuracies in the intercomparisons. Therefore no attempt to alter the lidar repeatabilities is made here.

2.2. Ozonesondes

In ozonesondes, ambient air is pumped through an electrochemical cell containing a buffered KI solution, in which a reaction is invoked by the ozone molecules [*Komhyr et al.*, 1995]. The calibration and operational procedures concerning the Lauder sondes are described by *Boyd et al.* [1998]. All sondes were manufactured by the Environmental Science Corporation. Two kinds of ozonesondes were flown within the time interval considered here; they can be distinguished by a difference in the sonde cathode solution. From August 1, 1996, onward, the cathode solution was diluted from 1% to 0.5% KI, which also included a halving of the buffer concentration. The 0.5% solution sondes are found to suffer less from "hysteresis" above the maximum ozone density and are therefore expected to yield more reliable results above ~22 km [*Boyd et al.*, 1998]. The sonde accuracy was derived from a Monte Carlo analysis and is better than 5% in the troposphere, 2-4% in the stratosphere up to about 30 km, and 4-7.5% between 30 and 35 km [*Bodeker et al.*, 1998].

2.3. Microwave Radiometer

The microwave radiometer detects the emission of radiation by ozone molecules in the gigahertz range. The ozone mixing ratios are derived, as a function of pressure, from the shape of the emission lines. To facilitate comparison with the other instruments, the pressure scale of the data has been converted into geometric altitude using NCEP temperature and pressure profiles for Lauder and the ideal gas law. In the present work, ozone profiles measured by the Millitech-NASA Langley microwave radiometer [*Parrish et al.*, 1988, 1992] are included. This radiometer has been operational at Lauder since 1992, with an intermission from late 1994 until March 1995.

The repeatabilities of the microwave radiometer ozone number densities are reported to be 4-5% throughout the altitude range, while the accuracies are 5-7% [*Con-*

nor et al., 1995; *Tsou et al.*, 1995]. As the microwave radiometer data are currently being revised, only a subset of the data is available for presentation here. They are ozone profiles obtained between April 1995 and January 1998, within 4 hours from lidar measurements. A paper describing in detail the revisions made and their validation is in preparation (J. J. Tsou et al., to be submitted to the *Journal of Geophysical Research*).

2.4. SAGE II Instrument

SAGE II is a limb-scanning Sun photometer, aboard the Earth Radiation Budget Satellite, which has been operated since October 1984. Height profiles of several atmospheric constituents, like ozone, NO_2, and aerosols, are retrieved from solar occultation measurements in seven wavelength bands between 0.385 and 1.02 μm. Calibration is achieved by comparing with measurements of radiation traveling through the exosphere. The ozone concentrations, inferred from the 0.6-μm channel, have an accuracy of about 5% in the stratosphere, with a vertical correlation distance of about 3 km [*McCormick et al.*, 1989]. Below 16 km the errors in the retrieved ozone profiles increase rapidly to about 40% at 10 km. Version 5.96 SAGE II ozone profiles measured within 2.5° latitude (~280 km) and 12° longitude (~940 km) from Lauder are included in the present work.

2.5. Dobson Spectrophotometer

For the Dobson spectrophotometer 72, located at Lauder, several wavelength bands are selected in the range from 305.5 to 339.8 nm (the Dobson AD bands [*Komhyr et al.*, 1989]) to evaluate ozone column densities, expressed in Dobson units (DU) (1 DU = 2.69×10^{16} molecules cm^{-2}). Standard ozone absorption cross sections [*Paur and Bass*, 1985] are used, based on laboratory measurements. They apply to a standard atmosphere for 45° N and a standard ozone profile for a column density of 325 DU [e.g., *Komhyr*, 1980]. Inherent in these assumptions is a fixed atmospheric temperature profile. The accuracy of the Dobson technique for direct Sun observations (the technique with which all Dobson measurement results mentioned here were collected) is estimated to be about 3% [*Komhyr et al.*, 1989].

The Dobson spectrophotometer at Lauder, operated by the National Oceanic and Atmospheric Administration and the National Institute of Water and Atmospheric Research, was compared against the World Standard Dobson Instrument 83 in February 1997. It was found to compare well, showing a maximum difference of 0.4% for direct Sun observations in the airmass range 1.15 to 3.2 [*Boyd et al.*, 1998]. System descriptions can be found elsewhere [e.g., *Komhyr et al.*, 1989].

3. Intercomparison Method

Since an annual cycle is expected in the ozone profiles and measurements by different instruments are not similarly spaced throughout the year, multiyear averages of all available ozone profiles measured by each of the instruments cannot be intercompared directly. Instead, ozone profiles measured by different instruments are included in the comparison only when measured as a pair, defined as two measurements performed within 24 hours (or within 4 hours for comparisons with microwave results) of each other. This criterion is checked for each altitude individually. Since different altitude ranges may be covered in different measurements, the number of contributors to a paired average is a function of altitude. The described procedure results in multiyear averages of paired ozone profiles.

Although atmospheric changes may introduce differences between ozone profiles in a pair, it is assumed that such differences average out and that therefore paired averages are not affected by atmospheric changes. This assumption is valid if a sufficiently large number of pairs is considered and if atmospheric changes between paired measurements are not systematic. A diurnal cycle in the ozone densities is not expected to cause systematic differences, because above 10 km the expected day-night differences are small (with the possible exception of differences at the top of the stratosphere).

Differences between paired ozone profiles, arising from the sondes drifting downwind during ascent, while other ground-based instruments measure in the zenith direction, are usually small. This was verified by regularly performed consecutive lidar measurements within a night (each with an integration time of 30 min) generally yielding similar ozone profiles. Since the air is in flux and the ozone lifetime is relatively long, we conclude that the ozone content of air downwind from the lidar is similar to that overhead.

The choice of a 24-hour maximum between measurements is somewhat arbitrary. When the allowed time difference is decreased to a few hours, the intercomparison results do not change significantly, except for an increased uncertainty in the smaller sample due to the smaller number of measurements included. An increase to 48 hours does not change the main conclusions significantly, except at low altitudes, where the influence of atmospheric variability increases.

In section 4, results of the comparison of the paired averages of the ozone profiles are discussed for various time intervals, thereby accounting for instrument changes that took place. Using linear interpolation techniques, ozone profiles from each pair of instruments have been put onto uniform altitude grids, i.e. the coarsest altitude grid of the instrument pair. Sonde data, typically recorded at a resolution of about 30 m, were first averaged to a 300-m grid and then linearly interpolated to coarser altitude grids. The lidar grid, described in subsection 2.1, is mainly characterized by an altitude grid with a bin size of 300 m, although some ozone profiles with bin sizes of 100 or 150 m have been included. Microwave data have typical bin sizes

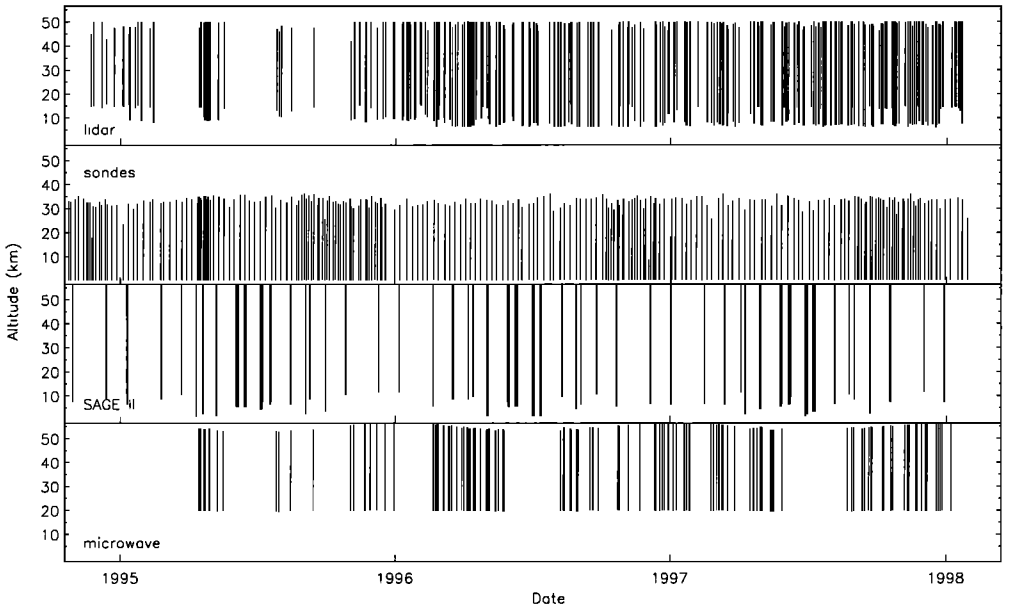

Figure 1. Overview of the time periods in which ozone profiles were measured at Lauder, New Zealand, ranging from November 1994 to January 1998. Each vertical line represents one measurement. The length of the lines gives the altitude range covered. From top to bottom, operational periods of lidar, sondes, SAGE II, and microwave radiometer are shown. Note that only the data used in this work have been plotted, which for the microwave radiometer is a subset of all measured data.

of about 2 km, while SAGE II data are presented on a 1-km altitude grid.

4. Comparison of the Ozone Profiles

An overview of the time periods of the measurements that are included in this work is shown in Figure 1. Clearly, measurements are not evenly spaced in time. There are several gaps in the data series, due to instrument problems. From the microwave radiometer, which in principle takes continuous measurements of 4-6 hours each, only a small subset of the revised data was available for this work. For the other instruments Figure 1 gives a fair indication of the number of ozone profiles measured per year: about 100 for the lidar (mainly determined by weather conditions), 60 for the sondes (twice weekly in spring and once weekly during the rest of the year), and 35 during SAGE II overpasses within 2.5° latitude and 12° longitude.

The intercomparison results are shown in subsection 4.1. We have compared paired averages of ozone profiles for all instrument combinations and different spans of time, but we will show only a selection of these results. The dates of the most recent hardware changes are 1996.22 (decimal year) for the lidar and 1996.58 for sondes. These dates have been taken as starting points for subsets of averages, as they best indicate the quality of the current ozone measurements at Lauder.

For a given time period, paired averages of ozone profiles are presented, along with accuracies and standard deviations within the set (see, for example, the left panels of Figures 2 and 3, which will be discussed in subsection 4.1). This standard deviation is a measure of profile-to-profile variability within the selected interval of time, which at most altitudes is dominated by atmospheric variability. Also the relative differences of the averaged profiles, along with the estimated repeatabilities in these relative differences, are calculated from the repeatabilities of the paired averages by using standard Gaussian error propagation (see, for example, the right panels of Figures 2 and 3).

The time dependence of the intercomparisons is also investigated, and for the interval between November 1994 and January 1998 no significant trends were found. However, it should be emphasized that the statistical uncertainty in this time interval is large.

4.1. Results

In Figure 2 paired averages (as discussed in section 3) of ozone profiles measured by lidar and by each of the other instruments are compared, for dates between November 1994 and January 1998. The top panels of Figure 2 show the lidar and sonde results. Evidently, the average profiles in the top panel are similar but appear to be offset in altitude. This is caused in part by the 50-m error in the lidar altitude scale discussed in subsection 2.1 (not corrected for in Figures 2 and 3). Another cause will be discussed in subsection 5.2. The relative difference (ozone densities measured by (lidar-sonde)/lidar) is within 4% above 20 km and within 10% between 12 and 20 km. This agrees with the results found during the OPAL campaign in April 1995 [*Mc-*

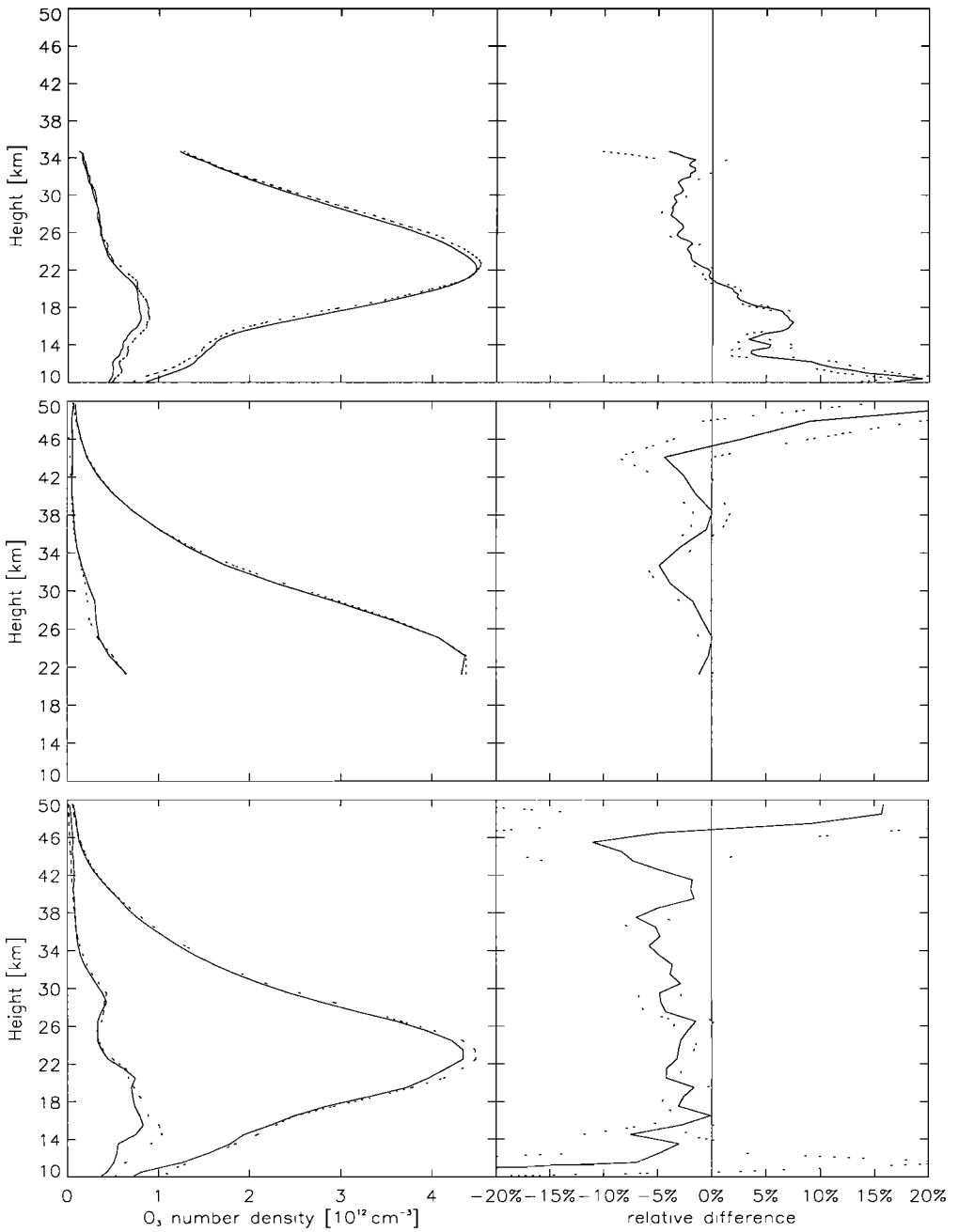

Figure 2. Averages of the lidar data (solid lines) compared with paired sonde (dashed lines, top), microwave (dashed lines, middle), and SAGE II (dashed lines, bottom) data for the November 1994 to January 1998 time period. In the three left panels, the average ozone number densities are shown versus altitude on the right side, along with the standard deviations in these averages on the left side. For clarity, the 2σ accuracies on these averages are not plotted, but up to about 30 km they are at least an order of magnitude smaller than the standard deviations. The three right panels show the differences between lidar data and paired sonde (top), microwave (middle), and SAGE II (bottom) data, relative to lidar, with the 2σ deviations of these relative differences (dashed lines).

Dermid et al., 1998a]. The relative differences below about 20 km are larger than those at higher altitudes, because of larger atmospheric variability and fewer contributing measurements. This makes the assumption that the same atmosphere is measured less valid. Instrument effects in the lidar and sonde data, which are discussed in section 5, also affect these differences. The number of pairs contributing to the lidar sonde comparison is about 110 (fewer at the top and bottom of the averaged profiles).

The middle panels of Figure 2 show the comparison of lidar and microwave results. Microwave ozone densities

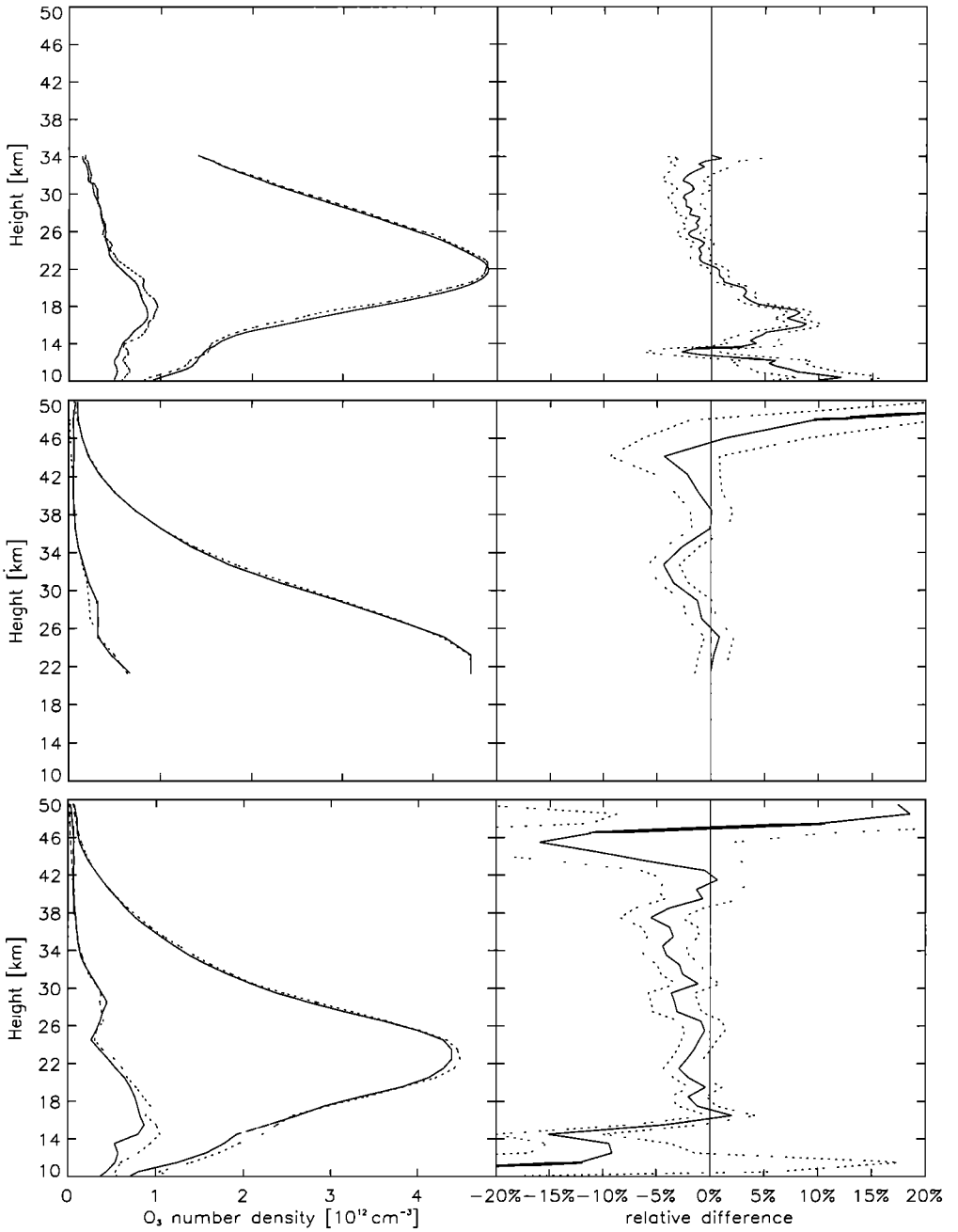

Figure 3. The same data as in Figure 2, but now for shorter time intervals: August 1996 to January 1998 for the comparison of lidar and sonde data (top panels) and March 1996 to January 1998 for the comparisons of lidar with microwave (middle panels) and lidar with SAGE II (bottom panels). Differences with Figure 2 reflect lidar and sonde hardware changes just before March 1996 and August 1996, respectively.

are larger than those of the lidar at all altitudes up to about 45 km, above which the lidar measurements are unreliable. The relative differences below 45 km range between 1% and 5%. The number of microwave/lidar pairs is about 180. Note that the microwave radiometer does not provide profiles at low altitudes.

The bottom panels of Figure 2 show the comparison of lidar and SAGE II (version 5.96) results. Clearly, at all altitudes below 48 km the SAGE II ozone densities are higher than those measured with lidar (see subsection 5.3). On average, the relative difference is about 3.5% between 20 and 40 km. Between 12 and 20 km the difference is between 5 and 8%. From comparisons with the ozone profiles measured by sondes and lidar, it is concluded that SAGE II results become increasingly less reliable with decreasing altitude. This is partly reflected in the large uncertainties given for the SAGE II data at low altitudes. SAGE II results at

low altitudes also compared poorly during the OPAL campaign [*McDermid et al.*, 1998a]. In a new version of the SAGE II data (SAGE II version 6, results to be published), in which a revised algorithm is used, the low-altitude results are expected to improve. The number of lidar/SAGE II and sonde/SAGE II pairs is 31.

The lidar ozone profiles appear to be unreliable above about 45 km, which follows from comparisons with SAGE II and microwave results. This may be due to the use of a numerical filter, which smoothes the lidar data over an interval of about 2.5 km. At high altitudes, this results in lidar ozone densities that are too high [*Godin et al.*, 1999]. Also the influence of background light and the background noise is a possible cause of inaccuracies at high altitudes.

Low-altitude lidar data recorded before March 1996 should be treated with caution (see subsection 2.1). For this reason, separate intercomparisons for the period between March 1996 and January 1998 (dates between 1996.22 and 1998.20) were made. For the intercomparison of sondes and other instruments, the interval of time was further reduced to start in August 1996 (dates between 1996.58 and 1998.20) so that it contains only the more accurate 0.5% KI solution sondes. There is a striking difference between the sonde/lidar comparison after 1996.58, shown in the top panel of Figure 3, and that for the whole period, shown in the top panel of Figure 2. In the shorter interval of time (Figure 3), the lidar and sonde ozone number densities agree within 2.5% for the 20- to 35-km altitude range (compared with 4% for the 3-year averages) and 8% for the 12- to 20-km altitude range (10% for the 3-year averages), respectively. In previous work [*Boyd et al.*, 1998] it was shown that at higher altitudes, lidar and sonde results agree better when 0.5% KI sondes are used than when 1% KI sondes are used, owing to reduced hysteresis in the 0.5% KI sondes, which agrees with the current results. The improvement seen between 12 and 15 km is not expected to stem from the changes in sonde hardware, since no significant changes are expected in sonde results below 15 km. Thus the lidar data were more reliable below 15 km after 1996.58 than they were before, which indicates that hardware problems might indeed have been affecting the lidar data in the older results.

The microwave/lidar intercomparison results, shown in the middle panels of Figure 3, are not significantly different before and after 1996.22. The relative difference between lidar and microwave ozone densities is within 5% for altitudes below 45 km. The number of contributing pairs is now about 75 for lidar/sonde, 140 for lidar/microwave, and 28 for lidar/SAGE II.

Finally, the SAGE II/lidar intercomparison, shown in the bottom panels of Figure 3, improved slightly after 1996.22 in the 20- to 35-km altitude range, agreeing to within 4.5%, which is almost within the combined accuracies (at the 2σ levels), although the SAGE II ozone number densities are still systematically higher by about 2.5% at all altitudes. Between 12 and 20 km, the agreement between lidar and SAGE II results has further deteriorated to up to 15%.

4.2. Influence of "Smoothing"

It is important to consider that differences between the results of measurements by various instruments could be caused in part by different altitude resolutions. Of the instruments considered, the sondes have the finest altitude resolution. They measure with a typical altitude spacing of about 30 m, but the resolution due to time needed for the chemical solution in the sondes to respond to ozone changes is about 150 to 200 m, typically. Before using them in the intercomparisons, the original sonde results have been averaged to a 300-m grid ("rebinned sonde profiles"). Lidar and SAGE II ozone profiles have resolutions of the order of a few kilometers, which is considerably coarser.

To investigate the influence of smoothing introduced by the lidar algorithm on the intercomparison, a simulation program was used. This program creates simulated lidar signals from high-resolution (30 m) sonde ozone profiles and then applies a numerical algorithm to these data, generating ozone profiles with an altitude resolution typical for the RIVM lidar results ("simulated lidar profiles").

These simulated lidar profiles are compared with the rebinned sonde profiles used in the intercomparisons (not shown). The simulated lidar profiles are smoother than the rebinned sonde profiles, while they were generated from identical original high-resolution data. This leads to a small systematic difference in ozone densities in the peak of the profiles (approximately between 20 and 25 km), where the ozone densities in the simulated profiles are consistently somewhat lower than those in the rebinned sonde profiles. No systematic altitude offsets were found. In conclusion, only small effects are expected if the difference in resolution between rebinned sonde profiles and lidar data is ignored. Therefore it is justified to use the rebinned sonde data for the intercomparisons.

Another effect of smoothing is present in the lidar profiles above about 40 km, where the poor resolution (of 2.4 km, corresponding to a 4.8-km interval; see subsection 2.1) used in the DIAL method, in combination with the shape of the ozone profile, introduces overestimates of the ozone densities, thereby biasing the lidar-SAGE II and lidar-microwave comparisons [*Godin et al.*, 1999].

The result of smoothing the sonde data with the SAGE II resolution has not been investigated, but since the resolutions of SAGE II and lidar are similar, it is expected that, also in this case, the use of rebinned sonde data is justified.

The microwave results have an altitude resolution of 7 to 15 km, which is coarser than that of the other results described. Based on the work of *Tsou et al.* [1995] for a

previous campaign, we conclude that the intercomparison will improve only slightly when the lidar profiles are smoothed to this resolution. Therefore the use of the original lidar ozone profiles in comparisons with the microwave data is justified.

5. Discussion and Proposed Modifications of Ozone Profile Analyses

The ozone profile intercomparison was discussed in section 4, where small discrepancies between the various results were found. In this section, possible causes for these discrepancies are discussed. This leads to proposed revisions of the ozone profiles. Where possible, the influence of these revisions is made quantitative. Intercomparisons of the modified data are presented in subsection 5.4.

5.1. Lidar Analysis

As was discussed in subsection 2.1, the altitudes used in the lidar results should be increased by 50 m. Motivated by the discrepancies between lidar and other results, we also critically reviewed the applied Rayleigh correction and ozone absorption cross section used in the RIVM lidar analysis. In this subsection, modifications to these parts of the analysis will be proposed. Subsequently, the resulting modified lidar data will be presented and compared with the version 7.3 lidar data shown in Figure 3.

Possible causes for the remaining discrepancies between lidar and other instruments were investigated, e.g., the high-count-rate nonlinearity of the detector and counting system and the applied correction for signal-induced noise [*Brinksma et al.*, 1997]. Since no evidence for these causes was found, they are not discussed further.

5.1.1. Rayleigh correction. The Rayleigh correction (the second term on the right-hand side of equation (1)) is proportional to the product of air density and the difference between the molecular Rayleigh extinction cross sections at 308 and 353 nm. The relative difference between lidar and sonde and between lidar and SAGE II (see Figure 3) ozone profiles may partly be explained by an incorrect Rayleigh correction. As was discussed by *Bucholtz* [1995], the Rayleigh extinction cross sections are underestimated in some publications by about 5%. (Note that the often cited paper by *Bates* [1984] contains values close to those presented by *Bucholtz* [1995].) In Table 1, the *Bucholtz* [1995] and old Rayleigh extinction cross sections are presented. We found that adapting the lidar Rayleigh corrections to incorporate the Bucholtz values led to decreases in the averaged lidar ozone densities of about 1% at 20 km, up to about 10% at 10 km, and significantly better agreement between lidar and sonde results.

In the modified lidar data, these Bucholtz values will be used. Also the sonde temperature and density profiles employed, typically recorded within 3 to 15 days from the lidar measurements, will be replaced by NCEP temperature and density profiles interpolated to values within a few hours of the lidar measurements. In a future version of the lidar data, either the sonde data closest in time (usually within 3 days from the lidar measurement) or NCEP data will be used, but those data will be very close to the modified lidar data presented below.

Table 1. Rayleigh Cross Sections Used in the Modified and Original Lidar Results

λ, nm	σ_{Ray} Modified, 10^{-26} cm^{-2}	σ_{Ray} Original, 10^{-26} cm^{-2}
308	5.03	4.88
353	2.82	2.74

Modified values from *Bucholtz* [1995].

5.1.2. Temperature dependence of the ozone absorption cross section. The altitude profile of the ozone absorption cross section, for which a temperature dependent expression must be used [*Paur and Bass*, 1985], is deduced from sonde measurements of the temperature. In the current lidar algorithm, the atmosphere is assumed to be isothermal above the sonde burst altitude (which is typically between 30 and 35 km). This introduces underestimates of the ozone absorption cross section at larger altitudes. In a future version of the RIVM lidar algorithm, truly simultaneous temperature profiles derived from the lidar signals themselves will be used, but this method is not yet operational. Therefore a modified data set was created in which NCEP temperature profiles (interpolated to values for the Lauder coordinates and within a few hours from the lidar measurements) were used to determine the ozone cross sections. This decreased the ozone number densities over the March 1996 to January 1998 interval of time by 1% at 35 km and up to 4% at 50 km.

5.1.3. Modified lidar ozone profiles. The modifications discussed above were applied to the lidar ozone profiles. In Figure 4, an intercomparison is shown between these modified profiles and the original data. Three altitude regions in which the lidar data have changed can be distinguished. At low altitudes, the modified Rayleigh corrections decrease the ozone number densities by 10% at 10 km, 4% at 15 km, and less than 1% above 20 km, on average. In the main part of the stratosphere, between 20 and 35 km, changes smaller than 1% are found, which are a combination of all three modifications which have been applied. Above 35 km, the ozone number densities decrease by 2% at 40 km and up to 5% at 50 km owing to the use of more realistic temperature profiles. The effect of the altitude increase by 50 m is very small.

The comparisons between modified ozone profiles measured by different instruments will be shown in subsection 5.4.

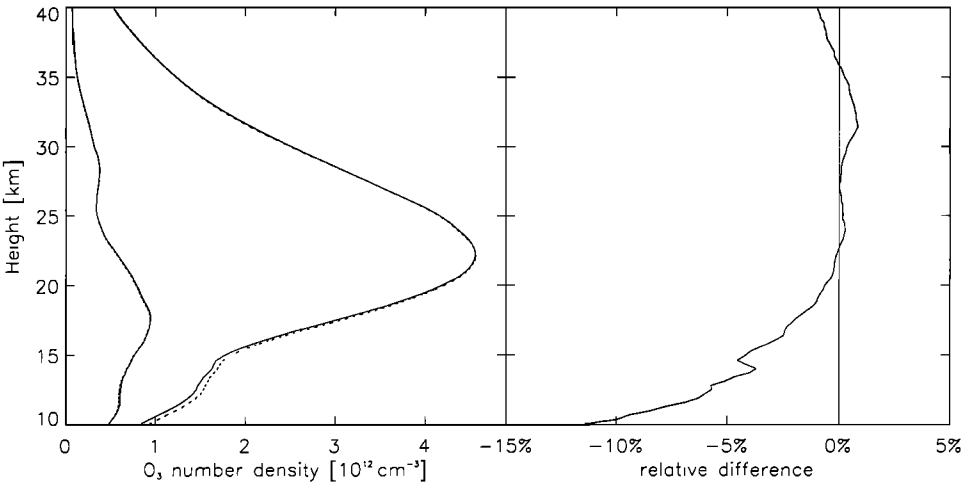

Figure 4. (left) Averages of the original lidar profiles (dashed lines) and the modified lidar profiles (solid lines) measured between March 1996 and January 1998 are shown on the right side, with the standard deviations in the averages on the left side. In the modified data, three changes have been applied, namely a 50-m increase of the altitudes, a change in the Rayleigh corrections and the implementation of a more realistic temperature dependence of the ozone cross section above about 35 km. (right) Relative difference between the averages, relative to the average of the modified data.

5.2. Ozonesonde Analysis

It was realized recently that most types of ozonesondes, including the ECC sondes used at Lauder, suffer from a time lag in the registration of ozone which is described by an *e*-folding response time of about 20 s (see, e.g., the *SPARC* [1998] report). This time lag causes an overestimate of the altitudes in the sonde ozone profiles. Common practice has been to ignore this altitude offset, but currently this is a topic of discussion. For the Lauder sondes, the response time varies between individual sondes and is on the order of 20 seconds, for both 0.5 and 1% KI sondes [*Boyd et al.*, 1998]. An assessment of the influence of an exponential response time on the ozone profile was carried out by using a computer simulation. A constant response time and constant sonde ascent rate were assumed. We found that a response time of 20 seconds yields an overestimation of the altitudes by about 150 m.

We applied several altitude offsets to the sonde ozone profiles and compared these profiles to (modified) lidar and SAGE II data. An offset of 125 m (±50 m) was found to give the best comparison between sondes and lidar, which is shown in Figure 5. Note that the agreement between lidar and sonde profiles has improved considerably in comparison with original results (which were shown in Figure 3, top panel). In particu-

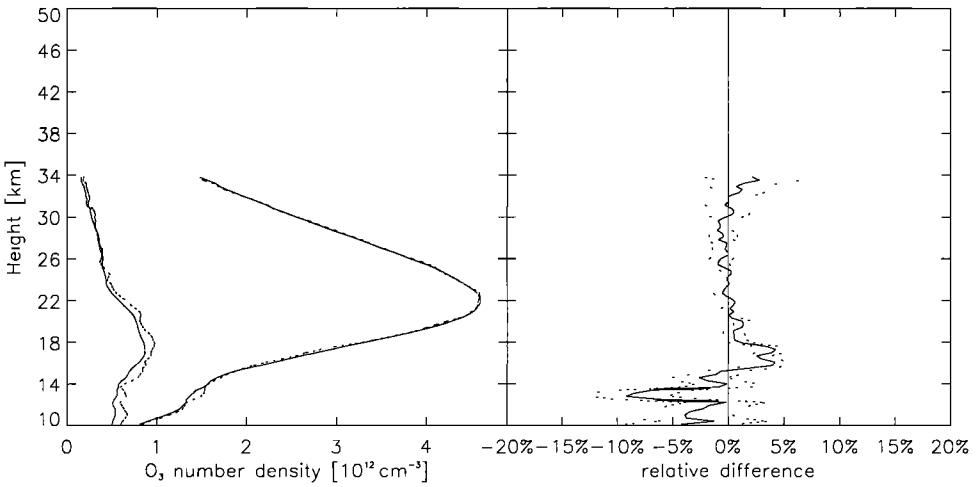

Figure 5. As in Figure 3, top panel, but with modified sonde data (dashed lines), in which the altitudes have been decreased by 125 m in comparison with the ozone profiles presented in Figure 3, and modified lidar data (as shown in Figure 4.).

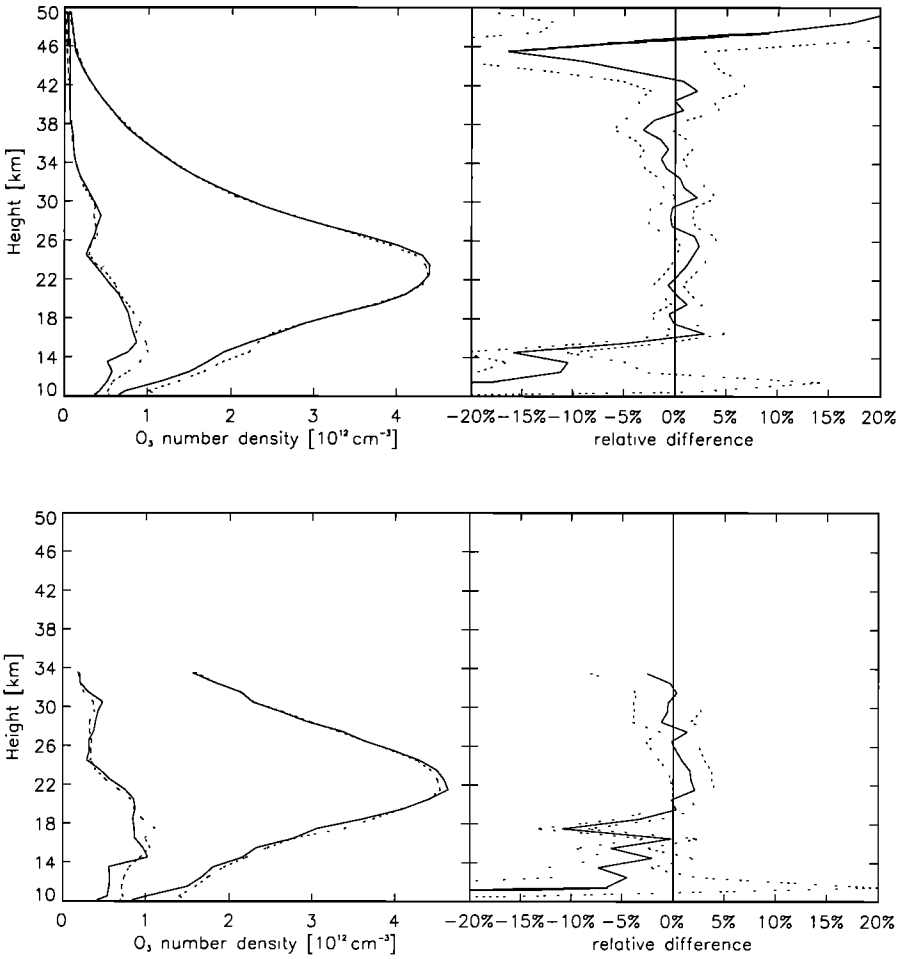

Figure 6. In the left panels, averages of the paired SAGE II (dashed line) and lidar (solid line, top) data and of the paired SAGE II (dashed line) and sonde (solid line, bottom) data are shown, along with the standard deviations in these averages. In the right panels, the relative differences between these averages, relative to the averages of lidar (top) or sonde (bottom), are shown. In both figures, the original SAGE II data have been multiplied by a factor of 0.975 at all altitudes. Lidar data have been modified as shown in Figure 4; sonde altitudes were decreased by 125 m. Data shown were obtained from measurements between March 1996 and January 1998 (lidar/SAGE II) or between August 1996 and January 1998 (sonde/SAGE II).

lar, the ozone densities agree within 1.5% in the main part of the stratosphere (20-35 km). This accuracy margin is small in comparison with the natural variations in ozone observed during the time period over which the average was made (i.e., the variance shown in the left panel of Figure 5). Only the sonde data after 1996.58 (which contained a 0.5% KI solution) were used here, because the 1% KI solution sondes were less accurate above about 22 km.

The comparison with SAGE II does not contradict the assumption that the sonde altitude scale should be altered, but the magnitude of the sonde offset cannot be determined accurately. After modifications of the SAGE II data, which will be discussed in subsection 5.3, a comparison of modified sonde and SAGE II profiles will be shown.

Supporting evidence for the proposed altitude offset followed from a study of profiles of ozone and water vapor mixing ratios, measured by the same sondes. It was found that tropospheric layers of ozone-rich air were shifted by about 125 m from layers of water vapor poor air, which for physical reasons were expected to be at the same altitude (E. J. Brinksma, manuscript in preparation). This confirms that the altitude error is specific for the ozone sensor and that it was not caused by any potential error source that would affect the altitude scale of profiles of ozone and humidity equally, like, for instance, a systematic error in the pressure-temperature measurement.

5.3. SAGE II Analysis

From the comparison with lidar and with the 0.5% KI altitude-corrected ozonesonde data, it is evident that the ozone density derived from SAGE II measurements is too large at all altitudes. This discrepancy is sig-

nificant with respect to the combined instrument accuracies. We have multiplied the SAGE II ozone densities with various numerical factors and found that best agreement of the SAGE II and lidar averages was found for a multiplication factor of 0.975 (±0.007). In Figure 6, data of lidar (modified as described above) and SAGE II (top panel) and of sondes and SAGE II (bottom panel) are shown in which the SAGE II ozone densities were multiplied by this factor of 0.975. The results indicate that an altitude-independent parameter in the SAGE II algorithm may be incorrect, e.g., the ozone absorption cross section may be underestimated. Indeed, in a future revision of the SAGE II algorithm (version 6) an ozone absorption cross section which is about 3% larger than the one currently used will be incorporated. This change is due to a different approach toward the combination of the extinctions at different wavelengths employed by SAGE II (J. M. Zawodny, private communication, 1999), and agrees with the multiplication factor we obtained.

5.4. Modified Intercomparison Results

We have proposed changes in the ozone profiles in three altitude regions. The effects of these changes on the intercomparison results are shown in this subsection.

Throughout the main part of the stratosphere, between 20 and 35 km, lidar and sonde results, modified as discussed above, agree to within 1.5% (Figure 5). The SAGE II data were multiplied by a factor of 0.975 and agreed to within 2.5% with modified lidar averaged profiles (1996.22-1998.1, 20-35 km, Figure 6, top panel) and with averaged modified sonde profiles (1996.58-1998.1, 20-33 km, Figure 6, bottom panel).

Between 12 and 20 km, modified lidar and sonde results are found to agree to within 9% (1996.58-1998.1). The SAGE II data in this altitude region are unreliable, and microwave data are not available at low heights.

Above 35 km, where a more realistic temperature dependence was implemented in the lidar ozone absorption cross section, the agreement between the microwave and lidar results has deteriorated slightly to 6% maximally (previously 5%; see Figure 3).

6. Validation of the Dobson Spectrophotometer at Lauder

In the previous section, ozone profiles measured with various instruments were compared. In this section, a fifth independent instrument, the Dobson spectrophotometer at Lauder, is considered, which measures ozone column densities. For those dates on which measurements by sondes and one of the other instruments were performed within 24 hours from the Dobson measurement, ozone column densities were also derived by integration with respect to altitude of the measured profiles. In order to cover the altitude region from ground level up to the top of the ozone layer, measured profiles

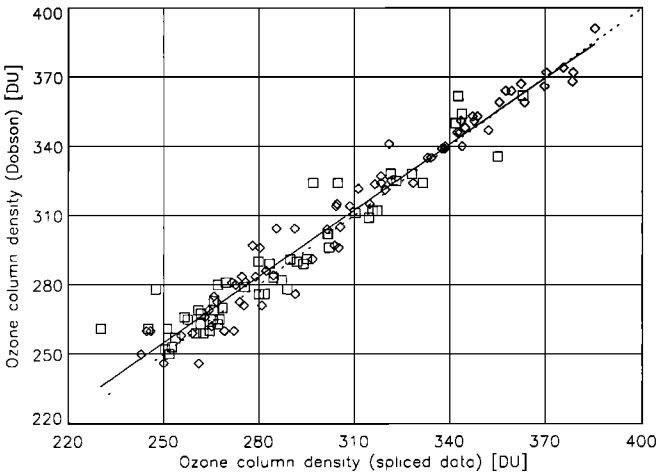

Figure 7. Scatter plots of the ozone column densities in Dobson units derived from the altitude integration of spliced sonde and lidar ozone profile measurements (horizontal axis) and from the paired Dobson measurements recorded within 24 hours from the spliced measurements (vertical axis). The spliced results were generated from modified sonde results below 20 km and modified lidar results above 20 km; 1% (squares) and 0.5% (diamonds) KI solution sondes are distinguished. The solid line represents a linear fit through all presented measurements. For reference, the dashed line of equality is also plotted.

are spliced in such a way that they consist of sonde data below a certain altitude (the "splice altitude") and data from one of the other instruments above that altitude.

The comparison of ozone column densities measured by the Dobson spectrophotometer and derived from sonde-lidar profiles, spliced at 20 km, is shown in Figure 7. Modified sonde and lidar data (see subsection 5.4) were used. Clearly, the overall agreement is good. A measure for the quality of the comparison is the root-mean-square (rms) of the differences in ozone column density determined by the Dobson and integrated spliced profile. This rms is 10 DU, corresponding to about 3% of the average ozone column density. When uncorrected sonde data are used, the rms difference is 9 DU. The splice altitude was also varied, to compare the sonde ozone column densities between 20 (or 15) and 30 km to the density derived from the other ozone profiling instruments, but this did not result in different conclusions.

Close inspection of Figure 7 shows that in the Dobson spectrophotometer results, high ozone column density values are underestimated, while low values are overestimated, in comparison with the integrated profiles. This is illustrated by fitting a straight line through all data shown. The slope of this fit is 0.95 (±0.04, 2σ accuracy), which is significantly different from slope 1.00.

Several parameters in the Dobson algorithm could introduce such a slope. Examples are the assumed altitude-averaged ozone absorption cross section, the assumed altitude distribution of ozone, and the solar

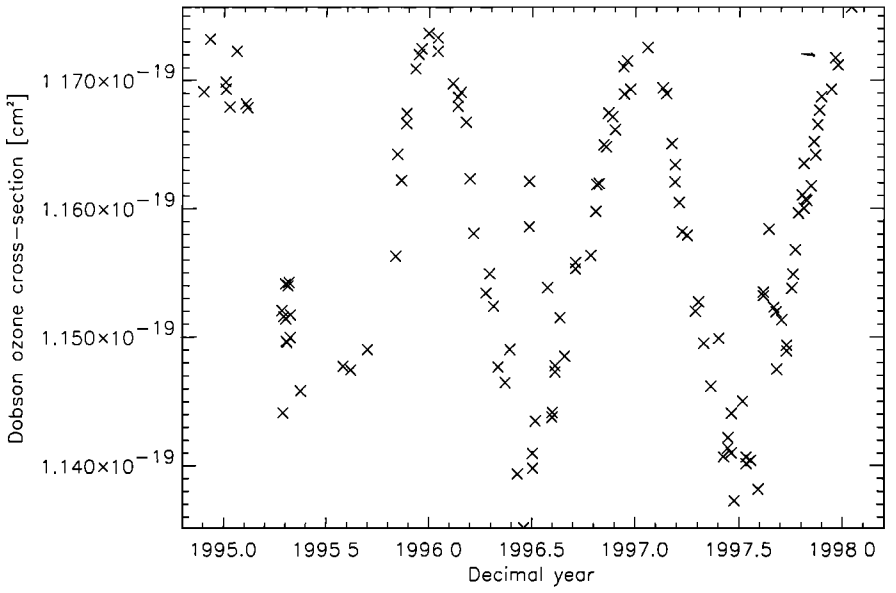

Figure 8. The Dobson ozone cross section as a function of time, derived from incorporating the NCEP temperature profiles in the Bass and Paur formulae and averaging the ozone cross section profiles with respect to altitude, weighted with the modified sonde and modified lidar ozone densities.

zenith angle at which Dobson observations are made. Only the first of these possible causes is investigated here.

Although it was noted previously [e.g., *Komhyr et al.*, 1989] that the weighted altitude average of the ozone cross section in the Dobson AD band (hereinafter, Dobson ozone cross section) is subject to an annual variation, often a fixed standard value, described in subsection 2.5, is used throughout the year. In the Dobson data discussed here, this was also the case. The influence of the assumption that the Dobson ozone cross section is constant can be made quantitative as follows.

The Dobson ozone cross section is calculated by first deriving altitude profiles of the ozone cross section in the Dobson AD wavelength bands [e.g., *Komhyr et al.*, 1989] from the Bass and Paur formulae [*Paur and Bass*, 1985]. These involve the temperature, for which the NCEP temperature profiles over Lauder are used. Second, weighted averages over all altitudes of the ozone cross sections in the AD band are made, in which the assigned weights are proportional to the ozone number densities from spliced sonde and lidar profiles. It is assumed that the four wavelength bands which characterize the AD band are infinitesimally small, and no correction is made for a difference in instrument sensitivity between these wavelengths. Also it is assumed that the Dobson ozone cross sections for a slant and straight column are identical.

The results of these two steps are displayed in Figure 8, in which the annual variation of the Dobson ozone cross section is shown, evidently with an amplitude of about 2%. This indicates that by assuming the Dobson ozone cross section to be constant, an error of the same magnitude is introduced. However, when the temperature-dependent Dobson ozone cross section is plotted against the observed ozone column densities or against the difference between integrated ozone profiles and Dobson results, no correlation is found. Thus the slope that was derived from Figure 7 cannot be explained by the annual variation of the Dobson ozone cross section. However, the results discussed are potentially important for future ozone column density intercomparisons at Lauder, because an error of up to 2% is introduced when the annual variation in the ozone absorption cross section is neglected.

7. Conclusions

This paper provides a representative validation of ozone profile data measured by various instruments, namely, all ground-based ozone profiling instruments at Lauder (New Zealand) and SAGE II. Before modifications were made, averages of lidar and sonde ozone number densities between 20 and 35 km agreed within 2.5% (August 1996 until January 1998), while averages of lidar, microwave and SAGE II ozone profiles agreed within 5% (March 1996 until January 1998).

In addition, the Lauder Dobson spectrophotometer was validated by using sonde and lidar ozone profiles. Agreement within 3% (rms difference) between ozone column densities derived from spliced sonde and lidar data and those measured by the Dobson spectrophotometer was found. Indications were obtained that the Dobson spectrophotometer underestimated high values of the ozone column density and overestimated low values of the ozone column density.

Based on NCEP temperature data and lidar and sonde ozone profiles, the Dobson ozone cross section (defined as the weighted altitude average of the ozone cross section in the Dobson AD band) is subject to an annual variation with an amplitude of about 2%. If the Dobson ozone cross section is assumed to be constant throughout the year, which is the case not only in this work but also in some other papers, an error is introduced, which over Lauder is up to 2% for the interval of time considered (December 1994 until January 1998).

Modifications of the algorithms for lidar, sondes and SAGE II are proposed. In the lidar algorithms, they encompass improvement of the determination of the Rayleigh correction and a more realistic assumption about the atmospheric temperature in the high stratosphere. In the sonde results, an altitude decrease of 125 m (±50 m) is proposed, to correct for the response time of the chemical solution to variations in ozone. It is important that the magnitude of this altitude offset will be determined in an independent manner (e.g., through laboratory measurements of the sonde response time) in future work. The SAGE II ozone number densities are found to be about 2.5% higher than those measured by lidar and 0.5% KI solution sondes at all altitudes. This could point to an inaccurate ozone absorption cross section, and indeed it was decided for other reasons than those described in subsection 5.3 that in a future version of the SAGE II data (version 6) the ozone cross sections will be increased by about 3%.

The agreement between lidar, sonde and SAGE II data, revised according to the proposed modifications, was excellent: averaged lidar and sonde ozone profiles agreed within 1.5% (20-35 km), while averaged lidar and SAGE II profiles agreed within 2.5% (20-35 km). Between 12 and 20 km the deviation between averaged lidar and sonde profiles became smaller than 9%. Microwave results were not revised.

As the validations show that the various techniques for measuring ozone profiles yield consistent results, it is useful to combine the various ozone profile measurements. The discussed instruments are complementary: only from the sonde results can tropospheric ozone profiles be derived, while SAGE II and the microwave radiometer are the only instruments providing reliable results above about 45 km. Between about 20 and 35 km, all of the instruments discussed measure mutually consistent data. This paper has shown that the apparent redundancy in this altitude region is useful, because it facilitates intercomparisons through which it is possible to find genuine mistakes, and their causes, and therefore to reach even better results by revising the data sets. Arguments for combining the data sets arise from the differences in temporal coverage and resolution of the various instruments. For future work it is potentially important to have a denser set of measurements spanning a larger altitude domain than a single instrument can provide.

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